Leveraging the Performance of Washing Cycles to Enhance the Preprocessing of Coal Fly Ash

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Abstract
Coal is relatively cheap and abundant fossil fuel contributing to 40% of the world's electricity production. However, coal combustion generates a significant number of by-products, majority being the coal fly ash (CFA). The heterogeneous nature of CFA poses significant challenges in its value-added applications hence, requiring appropriate preprocessing treatments which are expensive and technically savvy. Therefore, this research aims to improve CFA's suitability for the application of value-added products through a technique called “washing cycles” which wash the CFA multiple times using distilled water under a controlled environment with continuous and vigilant monitoring to ensure accuracy throughout the entire process. To optimise this washing cycle process, parameters such as the number of washing cycles, stirring time, and temperature were analysed, and the results were optimised through surface response methodology. The analytical findings on the optimisation process demonstrated that a suitable pre-treatment involved employing five washing cycles with a stirring time of 7.5 minutes. Furthermore, varying the temperature did not yield a substantial effect on the effectiveness of washing cycles, since the precipitation of Ca^{2+} ions even complicate the preprocessing. Therefore, we recommend conducting additional investigations into the efficient use of pre-processed CFA to produce value-added applications adhering to circular economy and sustainability.

Keywords: Coal fly ash, Conductivity, Response surface methodology, Washing cycles

1 Introduction

Demand for the coal used in electricity production reached its zenith in 2022, with consumption exceeding 8.3 billion tonnes [1]. The primary reason behind this trend is the prevalence of global economic challenges. In light of this, coal serves as an economically viable solution for electricity production due to its abundant availability in numerous parts of the world [1]. However, the disposal of CFA a by-product of coal combustion, has become a major environmental concern for countries heavily reliant on coal for power generation. Despite the many advantages coal provides, its combustion in thermal power plants poses significant risks to both the biotic and abiotic environments in their vicinity. CFA exposure can lead to various risks such as DNA damage, the heightened incidence of cardiovascular and respiratory ailments, and adverse effects on fatal development [2]. CFA constitutes a substantial proportion of ash produced, ranging from 65% to 95%, and consists of various mineral compounds, including oxides like SiO_2, Al_2O_3, CaO, Fe_2O_3, Na_2O, K_2O, MgO, TiO_2, and other metal oxides [3], [4]. Furthermore, CFA contains elevated levels of hazardous heavy metals such as, As, Cd, Pb, and Hg which can cause both human and aquatic animals as well [2]. Improper disposal of CFA can lead to environmental risks, such as water and soil contamination, and contribute to air pollution through the formation of smog and haze [5]. If CFA is disposed of for an extended period, it can worsen the problem of heavy metals as persistent pollutants. These metals do not break down naturally and can easily build up in living beings, even in small amounts, leading to severe health issues [3]. The search for suitable disposal sites, high maintenance costs of land management, and adverse environmental impacts discourage conventional disposal methods such as holding ponds, lagoons, landfills, and slag heaps-sentence and reuse methods [6]. Properly managing the disposal of CFA is crucial to avoid environmental harm, which can also result in financial liabilities. Therefore, a prudent approach is to transform this waste product into a value-added resource. CFA is primarily utilised in the cement industry, wastewater treatment, the agriculture sector, land reclamation activities, and so on [7]. There is a growing interest in recycling CFA into value-added products to address these issues and to promote eco-friendly practices and sustainable solutions [8]. Fly ash is widely used directly in various applications, encompassing agricultural soil enhancement, construction of roadways and pavements, manufacturing of cement and concrete products, production of ceramic materials, soil and water treatment, and catalytic applications [7]. One promising application of CFA is in wastewater treatment due to its high content of Silicon (Si) and Aluminium (Al). These elements make it a valuable source for synthesizing zeolitic materials, which find extensive use in wastewater treatment processes [9]. Zeolites are versatile and fascinating materials that are vital for a wide range of industries, due to their unique structural and chemical properties, which are the basis of applications in gas separation, ion exchange, and catalysis [10]. It possesses an inherent negative charge and features a cage-like structure. Zeolites are distinguished by their prominent characteristics of cation exchange and the absorption of inorganic and organic molecules with specific dimensions [11]. There are various methods to synthesise zeolites. Among various synthesis methods, the microwave-assisted hydrothermal approach stands out for its efficiency, rapidity, cost-effectiveness, and environmental friendliness [12]. In this approach, microwave energy is applied to the reaction mixture containing coal fly ash to accelerate the dissolution process, subsequently followed by crystallization through hydrothermal heating [13]. Careful control and optimisation of factors such as the origin of alumina and silica sources, Si to Al ratio, alkali solution to CFA weight ratio, synthesis method, and reaction conditions are crucial in determining the characteristics and properties of the produced synthesised zeolites [13]. The utilisation of CFA for wastewater treatment presents an opportunity to address waste management and water quality concerns [14].

CFA, a well-known waste material, is being utilised as a raw material in the treatment of wastewater [11]. Before synthesizing zeolite using CFA, it is essential to perform preprocessing to eliminate impurities, ensuring a higher yield of zeolite [12]. Among various preprocessing techniques, employing washing cycles stands out as one of the most efficient and budget-friendly methods [4], [15]. Additionally, it offers other benefits in terms of simplifying the complexity of CFA, reducing heterogeneity, and enhancing the overall quality for subsequent applications [13]. This study aimed to optimise the stirring duration, temperature, and the number of washing cycles to enhance zeolite effectiveness while minimising costs and energy consumption.

As there are limited discussions on optimising the preprocessing techniques related to washing cycles, this research sought to explore and determine the ideal parameters for achieving the desired outcomes.

2 Methodology

Because of the heterogeneous nature of CFA, consistent outcomes are not observed in every trial. Hence, it is crucial to simplify the complexity of CFA before employing it for any purpose. Considering the aforementioned challenge, the primary preprocessing method suggested to address the heterogeneity of CFA is washing it with distilled water [4], [15]. To improve the washing cycle process, several parameters can be optimised, including temperature, solid-to-liquid ratio, stirring speed, stirring time, and the number of washing cycles. In our study, we investigated how the effectiveness of washing cycles is influenced by the number of washing cycles, stirring time, and temperature. The conductivity of the solution is boosted by stirring, which leads to increased dissolution of readily soluble ions from the surface of the CFA particles. Experiments were conducted to understand the synergetic effect of stirring time, number of washing cycles, and stirring temperature in the process of washing cycles. Two experiment setups were designed for optimum time, number of washing cycles, and temperature determination. The experiments to assess the optimum stirring time and number of washing cycles were conducted by replacing distilled water after each washing cycle (i.e., replacement method) and without replacing distilled water after the end of each washing cycle (i.e., without replacement method).

The second experiment series was conducted to determine the optimum stirring temperature with the selected method, either with or without replacement.

2.1 Optimisation of stirring time and the number of washing cycles.

Optimum values for the stirring time and number of washing cycles were evaluated using both replacement and without replacement methods at different time intervals (5 min, 10 min, and 15 min) with 10-minute settling time at room temperature. Initially, two samples of 100 g CFA were mixed with 400 ml of distilled water in beakers separately. One beaker experimented with the replacement method and the other with the without replacement method. Five washing cycles were conducted under the replacement method and another five washing cycles were conducted under the without replacement method.

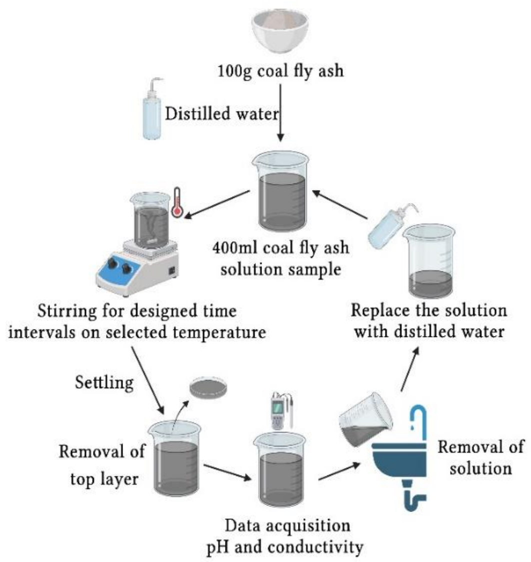

Figure 1: Experiment design for a single washing cycle

2.2 Data collection

Measurement collection was done after every washing cycle so that the conductivity of the solution was recorded. After each washing cycle, three conductivity measurements were recorded and subsequently averaged to obtain the arithmetic mean conductivity. The changes in conductivity reveal the degree of soluble ion dissolution in the solution and the effectiveness of impurity removal in each washing cycle. Analysing the conductivity concerning the number of washing cycles, stirring time, and temperature allows us to assess how these factors impact the removal of impurities. One washing cycle involves stirring at an appropriate time (5 min, 10 min, and 15 min), and 10 minutes of settling. The conductivity of the solution was measured using a Hach HQ40D digital conductivity meter at the end of each washing cycle. The top layer of the settled sample container was skimmed carefully before measuring the conductivity of the solution.

2.3 Optimisation of stirring temperature

Following the initial experiment with the previously optimised values, the stirred samples underwent 10-minute settling periods, consistent with the previous setup. Subsequently, a series of experiments were conducted at different temperatures: 30.5°C, 50°C, 60°C, and 70°C. The primary goal was to assess the influence of these varied temperatures on the settling process, with the ultimate objective of identifying the stirring temperature that would yield enhanced zeolite effectiveness while considering factors such as settling efficiency and energy consumption.

3 Results and discussion

Fig. 2 shows the conductivity trend with the increasing number of washing cycles and stirring time of the replacement method. The replacement method shows that conductivity has decreased with the increasing number of washing cycles. This phenomenon results from the discharge of ions (K^+, Na^+, Ca^+, and Mg^+) during every washing cycle with the replacement of the solution [13]. The maximum conductivity of the solution sample was 6.1600 mS/cm obtained after the first washing cycle of 15 minutes of stirring time.

In the without replacement method, Fig. 3 shows that conductivity has been increased with the increasing washing cycles and the difference between the conductivity of the solution with increasing the washing cycles has been decreased. The maximum conductivity of the solution sample was 6.0400 mS/cm, obtained after the third washing cycle of 15 minutes of stirring time.

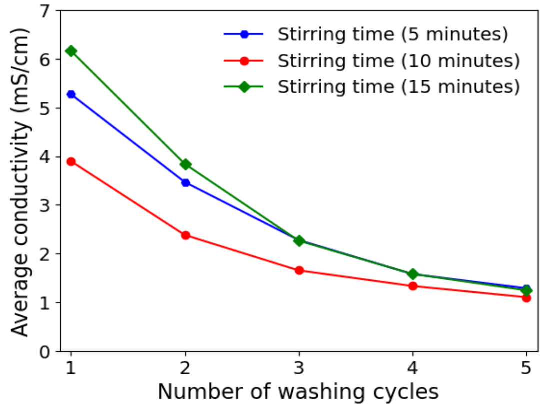

Figure 2: Variation in average conductivity with different washing cycles as influenced by temperature (replacement method)

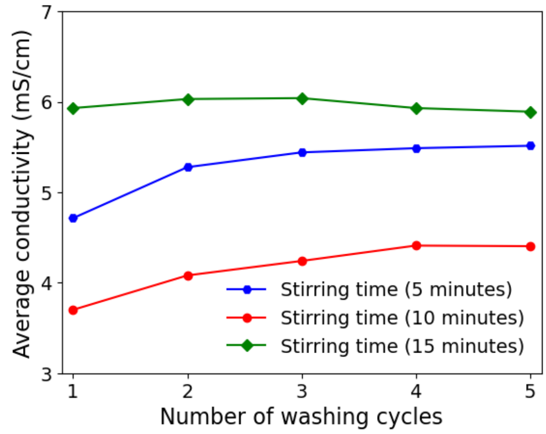

Figure 3: Variation in average conductivity with different washing cycles as influenced by stirring time (without replacement method)

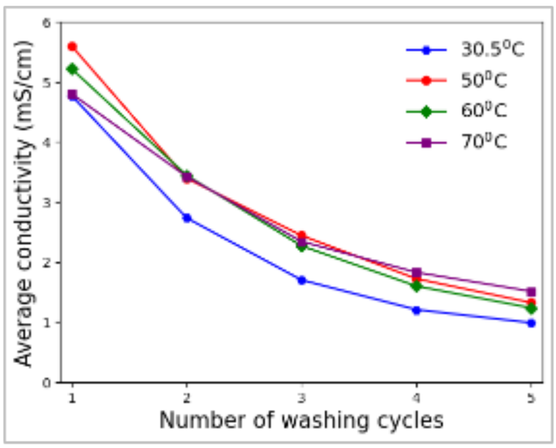

Figure 4: Variation in average conductivity with different washing cycles as influenced by stirring time (replacement method)

Further, the stirring temperature was optimised in the replacement method by keeping constant which is the stirring time (at 7.5 min), setting time, and other aforementioned factors. According to the obtained results, it can be concluded that the conductivities of the solution after each washing cycle at 30.5°C showed low conductivities when compared with the conductivity values at the temperatures of 50°C, 60°C, and 70°C. As the temperature increases, more Ca^{2+} ions dissolve in the solution. However, because of the presence of CO_3^{2-} and SO_4^{2-} ions, these Ca^{2+} ions tend to form solid precipitates such as $CaCO_3$ and $CaSO_4$ [13]. As a result, there is no significant change in conductivity with increasing temperature.

3.1 Numerical optimisation

The data processing for this study employed the utilisation of specialised software known as "Design Expert." The objective of the study was to optimise conductivity by assessing the possible range of variables for string time, the number of washing cycles, and the implementation of both replacement and without replacement methods. Minimising the conductivity obtained after a specific washing cycle was deemed an essential component in the optimisation process. The rationale behind this approach is that achieving the lowest conductivity after a given washing cycle signifies the highest level of ion dissolution in distilled water. Using the aforementioned concept and the level of desirability, the number of washing cycles was optimised.

The results obtained from Fig. 5 and Fig. 6 show that increasing the number of washing cycles results in maximum desirability and minimum conductivity. As desirability improves, it leads to enhanced accuracy and reduced conductivity, ensuring the maximum possible ion removal with an increased number of washing cycles.

The response surface of the conductivity optimisation analysis in Fig. 7 highlights that the minimum conductivity value is obtained when there are five washing cycles and a stirring time of 7.5 minutes while considering the categorical factor of using either the replacement or without replacement method. These findings suggest that these specific parameter values can result in improved performance and desired outcomes in the washing cycle phase of the experiment.

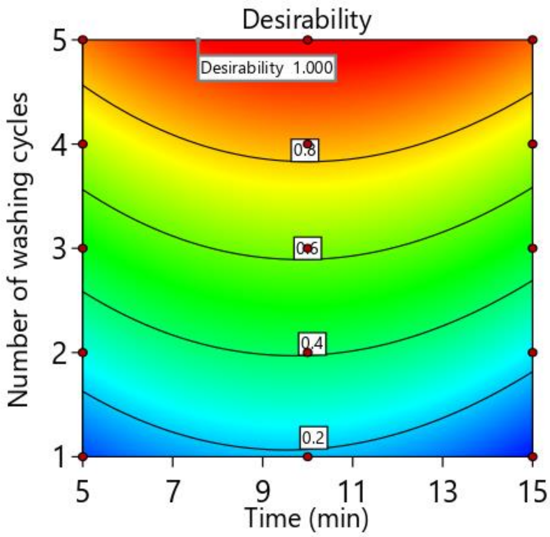

Figure 5: Contour plot of desirability

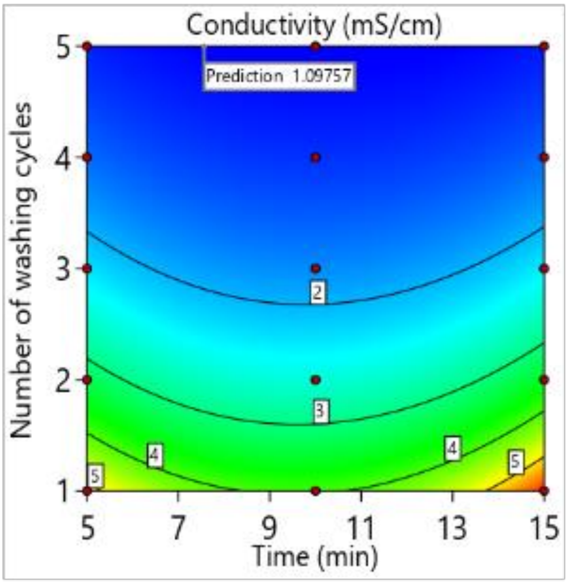

Figure 6: Contour plot of conductivity

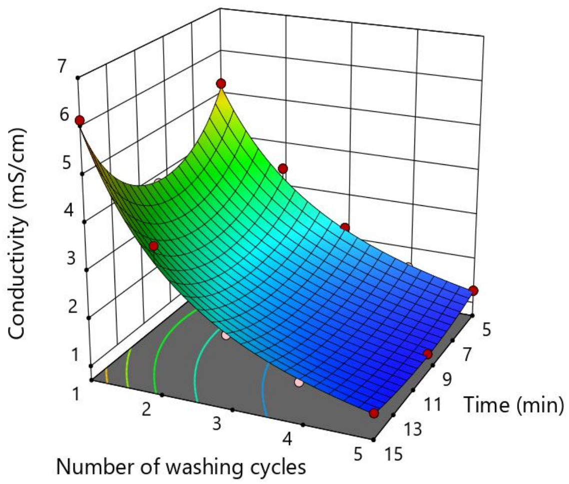

Figure 7: Response surface of the conductivity optimisation

5 Conclusion

Based on the conducted experiments, this research focused on optimising the washing cycles, and stirring time, and evaluating the impact of temperature on the conductivity of the solution. The results of the experiment indicate that five washing cycles are necessary to obtain the optimum conductivity of the solution. Furthermore, the study found that stirring the CFA solution for 7.5 minutes leads to optimum conductivity. These results highlight the importance of proper mixing and agitation in achieving the desired pre-processed CFA. Stirring the solution for a shorter or longer duration may result in suboptimal conductivity. In addition to that, the investigation into the effect of temperature on solution conductivity revealed that temperature changes did not have a significant impact on conductivity. The conductivity of the solution remained relatively stable across different temperature ranges in each washing cycle. This suggests that temperature variations may not be a crucial factor influencing the conductivity of the solution under the given experimental conditions.

These results offer valuable insights for future investigations and practical implementations of washing cycles as an effective preprocessing technique for CFA. Further investigation is needed to gain an in-depth understanding of various uncharacteristic behaviours of CFA. Specifically, assessing and optimising the solid-to-liquid ratio (CFA to distilled water) while considering other factors, like stirring rate, will be crucial in unravelling these complexities.

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References

[1] I. Energy Agency, “Coal Market Update_July 2023,” 2023. [Online]. Available: https://www.iea.org/

[2] H. Park, L. Wang, and J. H. Yun, “Coal beneficiation technology to reduce hazardous heavy metals in fly ash,” *J Hazard Mater*, vol. 416, Aug. 2021, doi: 10.1016/j.jhazmat.2021.125853.

[3] M. Visa and A. M. Chelaru, “Hydrothermally modified fly ash for heavy metals and dyes removal in advanced wastewater treatment,” *Appl Surf Sci*, vol. 303, pp. 14–22, Jun. 2014, doi: 10.1016/j.apsusc.2014.02.025.

[4] IE Pannilarathne, MGI Sandaruwan, P Mathissan, CL Jayawardena and WAM Fernando, “Characterisation of Coal Fly Ash for Potential Wastewater Treatment Opportunities,” Proceedings of International Symposium on Earth Resources Engineering and Management2021.

pp. 85–90, 2021. http://dl.lib.uom.lk/handle/123/17109

[5] N. Koshy and D. N. Singh, "Fly ash zeolites for water treatment applications," *Journal of Environmental Chemical Engineering*, vol. 4, no. 2. Elsevier Ltd, pp. 1460–1472, Jun. 01, 2016. doi: 10.1016/j.jece.2016.02.002.

[6] R. Iyer, "The surface chemistry of leaching coal fly ash," 2002

[7] C. Belviso, "State-of-the-art applications of fly ash from coal and biomass: A focus on zeolite synthesis processes and issues," *Progress in Energy and Combustion Science*, vol. 65. Elsevier Ltd, pp. 109–135, Mar. 01, 2018. doi: 10.1016/j.pecs.2017.10.004.

[8] Q. Xue, J. S. Li, P. Wang, L. Liu, and Z. Z. Li, "Removal of heavy metals from landfill leachate using municipal solid waste incineration fly ash as adsorbent," *Clean (Weinh)*, vol. 42, no. 11, pp. 1626–1631, Nov. 2014, doi: 10.1002/clen.201300619.

[9] T. V. Ojumu, P. W. Du Plessis, and L. F. Petrik, "Synthesis of zeolite A from coal fly ash using ultrasonic treatment - A replacement for fusion step," *Ultrason Sonochem*, vol. 31, pp. 342–349, Jul. 2016, doi: 10.1016/j.ultsonch.2016.01.016.

[10] V. Van Speybroeck, K. Hemelsoet, L. Joos, M. Waroquier, R. G. Bell, and C. R. A. Catlow, "Advances in theory and their application within the field of zeolite chemistry," *Chemical Society Reviews*, vol. 44, no. 20. Royal Society of Chemistry, pp. 7044–7111, Oct. 21, 2015. doi: 10.1039/c5cs00029g.

[11] S. K. Nayak and S. R. Mohanty, "Zeolite Synthesis from Waste and its Applications: A Retrospective," 2019. [Online]. Available: www.ijisrt.com

[12] S. Zeng, R. Wang, Z. Zhang, and S. Qiu, "Solventless green synthesis of sodalite zeolite using diatomite as silica source by a microwave heating technique," *Inorg Chem Commun*, vol. 70, pp. 168–171, Aug. 2016, doi: 10.1016/j.inoche.2016.06.013.

[13] B. Makgabutlane, L. N. Nthunya, E. N. Nxumalo, N. M. Musyoka, and S. D. Mhlanga, "Microwave Irradiation-Assisted Synthesis of Zeolites from Coal Fly Ash: An Optimization Study for a Sustainable and Efficient Production Process," *ACS Omega*, vol. 5, no. 39, pp. 25000–25008, Oct. 2020, doi: 10.1021/acsomega.0c00931.

[14] F. Mushtaq, M. Zahid, I. A. Bhatti, S. Nasir, and T. Hussain, "Possible applications of coal fly ash in wastewater treatment," *Journal of Environmental Management*, vol. 240. Academic Press, pp. 27–46, Jun. 15, 2019. doi: 10.1016/j.jenvman.2019.03.054.

[15] K Brinthan, S Shivadhahini, UAG Senadheera, WAM Fernando, CL Jayawardena, DRT Jayasundara, & MADMG Wickrama, "Experimental investigation and performance optimisation of washing cycles for pre-processing of coal fly ash". Proceedings of International Symposium on Earth Resources Engineering and Management 2022 pp. 70–77, 2022. doi.org/10.31705/iserme.2022.11